

FORM PTO-1390 (Modified) (REV 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER 219455US0PCT
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 10/069502
INTERNATIONAL APPLICATION NO. PCT/JP00/05844	INTERNATIONAL FILING DATE 30 AUGUST 2000		PRIORITY DATE CLAIMED 31 AUGUST 1999	
TITLE OF INVENTION METHOD OF MANUFACTURING 2,6-DIMETHYLNAPHTHALENE				
APPLICANT(S) FOR DO/EO/US Koji YAMAMOTO, et al.				
<p>Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:</p> <ul style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below. 4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ul style="list-style-type: none"> a. <input type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input checked="" type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ul style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ul style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). 11. <input type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). 12. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210). <p>Items 13 to 20 below concern document(s) or information included:</p> <ul style="list-style-type: none"> 13. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 15. <input checked="" type="checkbox"/> A FIRST preliminary amendment. 16. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 17. <input type="checkbox"/> A substitute specification. 18. <input type="checkbox"/> A change of power of attorney and/or address letter. 19. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 20. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4). 21. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 22. <input type="checkbox"/> Certificate of Mailing by Express Mail 23. <input checked="" type="checkbox"/> Other items or information: <p>Notice of Priority / PCT/IB/304 / PTO-1449 Drawings (3 sheets)</p>				

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 10/069502	INTERNATIONAL APPLICATION NO. PCT/JP00/05844	ATTORNEY'S DOCKET NUMBER 219455US0PCT
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24. The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO	\$1040.00
<input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO	\$890.00
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO	\$740.00
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)	\$710.00
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)	\$100.00

CALCULATIONS PTO USE ONLY

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$890.00

Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). 20 30

\$0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	16 - 20 =	0	x \$18.00	\$0.00
Independent claims	2 - 3 =	0	x \$84.00	\$0.00
Multiple Dependent Claims (check if applicable).			<input type="checkbox"/>	\$0.00

TOTAL OF ABOVE CALCULATIONS = **\$890.00**

Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.

\$0.00

SUBTOTAL = **\$890.00**

Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). 20 30 +

\$0.00

TOTAL NATIONAL FEE = **\$890.00**

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).

\$0.00

TOTAL FEES ENCLOSED = **\$890.00**

Amount to be: refunded	\$
charged	\$

- a. A check in the amount of \$890.00 to cover the above fees is enclosed.
- b. Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 15-0030. A duplicate copy of this sheet is enclosed.
- d. Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



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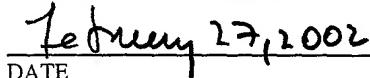

SIGNATURE

Norman F. Oblon

NAME

24,618

REGISTRATION NUMBER


DATE

DOCKET NO.: 219455 US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: Koji YAMAMOTO, et al.

SERIAL NO.: NEW U.S. PCT APPLICATION

FILED: HEREWITH

INTERNATIONAL APPLICATION NO.: PCT/JP00/05844

INTERNATIONAL FILING DATE: August 30, 2000

FOR: METHOD FOR MANUFACTURING 2,6-DIMETHYLNAPHTHALENE

**REQUEST FOR PRIORITY UNDER 35 U.S.C. 119
AND THE INTERNATIONAL CONVENTION**

Assistant Commissioner for Patents

Washington, D.C. 20231

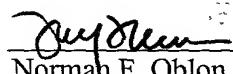
Sir:

In the matter of the above-identified application for patent, notice is hereby given that the applicant claims as priority:

<u>COUNTRY</u>	<u>APPLICATION NO</u>	<u>DAY/MONTH/YEAR</u>
Japan	11-246299	31 August 1999

Certified copies of the corresponding Convention application(s) were submitted to the International Bureau in PCT Application No. PCT/JP00/05844. Receipt of the certified copy(s) by the International Bureau in a timely manner under PCT Rule 17.1(a) has been acknowledged as evidenced by the attached PCT/IB/304.

Respectfully submitted,
OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.


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219455US-0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
KOJI YAMAMOTO ET AL : ATTN: APPLICATION DIVISION
SERIAL NO: NEW U.S. PCT APPLN :
(Based on PCT/JP00/05844)
FILED: HEREWITH :
FOR: METHOD FOR
MANUFACTURING
2,6-DIMETHYLNAPHTHALENE

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows.

IN THE CLAIMS

Please cancel Claims 1-9.

Please add the following new claims:

10. (New) A method for manufacturing highly pure 2,6-dimethylnaphthalene comprising: a step of performing cooling crystallization of a mixture containing dimethylnaphthalenes which includes 2,6-dimethylnaphthalene; a step of performing solid-liquid separation to obtain a solid component; and a washing step of washing the solid component using a solvent;

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wherein the solid-liquid separation performed after the cooling crystallization includes press filtration.

11. (New) The manufacturing method according to Claim 10, wherein the mixture containing dimethylnaphthalenes is a mixture composed of dimethylnaphthalene isomers.

12. (New) The manufacturing method according to Claim 10, wherein the press filtration is performed at a pressure of 10 kg /cm² or more.

13. (New) The manufacturing method according to Claim 10, wherein the mixture containing dimethylnaphthalenes used as a feedstock includes 5 wt% or more of 2,7-dimethylnaphthalene.

14. (New) The manufacturing method according to Claim 10, wherein the cooling crystallization is performed for a mixture containing dimethylnaphthalenes which includes less than 25 wt% of 2,6-dimethylnaphthalene.

15. (New) The manufacturing method according to Claim 10, wherein the washing step is performed for a solid component containing 80% or more of 2,6-dimethylnaphthalene using a solvent, and further comprising steps of performing solid-liquid separation and distillation after the washing step, whereby a 2,6-dimethylnaphthalene having a high purity of 99% or more is obtained.

16. (New) The manufacturing method according to Claim 10, wherein the solvent used in the washing step is an aliphatic hydrocarbon and/or alicyclic hydrocarbon having 5 to 10 carbon atoms.

17. (New) The manufacturing method according to Claim 10, wherein the press filtration is performed using a tube press.

18. (New) A method for manufacturing highly pure 2,6-dimethylnaphthalene comprising: a step of performing cooling crystallization of a mixture containing

dimethylnaphthalenes which includes 2,6-dimethylnaphthalene; a step of performing solid-liquid separation to obtain a solid component; and a washing step of washing the solid component using a solvent; wherein the washing step is performed at least twice, and a part or the entirety of a mother liquor obtained in a second washing step or in a subsequent washing step is used as a solvent in a washing step performed prior to the washing step at which the mother liquor is obtained.

19. (New) The manufacturing method according to Claim 17, wherein the mixture containing dimethylnaphthalenes is a mixture composed of dimethylnaphthalene isomers.

20. (New) The manufacturing method according to Claim 17, wherein the press filtration is performed at a pressure of 10 kg/cm² or more.

21. (New) The manufacturing method according to Claim 17, wherein the mixture containing dimethylnaphthalenes used as a feedstock includes 5 wt% or more of 2,7-dimethylnaphthalene.

22. (New) The manufacturing method according to Claim 17, wherein the cooling crystallization is performed for a mixture containing dimethylnaphthalenes which includes less than 25 wt% of 2,6-dimethylnaphthalene.

23. (New) The manufacturing method according to Claim 17, wherein the washing step is performed for a solid component containing 80% or more of 2,6-dimethylnaphthalene using a solvent, and further comprising steps of performing solid-liquid separation and distillation after the washing step, whereby a 2,6-dimethylnaphthalene having a high purity of 99% or more is obtained.

24. (New) The manufacturing method according to Claim 17, wherein the solvent used in the washing step is an aliphatic hydrocarbon and/or alicyclic hydrocarbon having 5 to 10 carbon atoms.

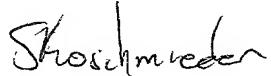
25. (New) The manufacturing method according to Claim 17, wherein the press filtration is performed using a tube press.

REMARKS

Claims 10-25 are active in the present application. Claims 1-9 have been cancelled. Support for the new claims is found in the original claims. No new matter is believed to have been added by this amendment. An action on the merits and allowance of claims is solicited.

Respectfully submitted,

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Marked-Up Copy

Serial No:

Amendment Filed on:

3/27/02

IN THE CLAIMS

Claims 1-9 have been cancelled.

Claims 10-25 have been newly added.

SEARCHED SERIALIZED INDEXED

- 1 -

DESCRIPTION

METHOD FOR MANUFACTURING 2,6-DIMETHYLNAPHTHALENE

Technical Field

The present invention relates to a method for manufacturing 2,6-dimetylnaphthalene which can be used effectively as, for example, a feedstock for 2,6-naphthalene dicarboxylic acid which is used for forming polyesters or the like.

Background Art

In order to obtain the superior features of polyethylene naphthalate which is used for manufacturing fibers, films, and the like, which are primarily formed of polyethylene naphthalate, it is necessary that 2,6-naphthalene dicarboxylic acid, which is a monomer component for forming a polymeric material, have a high purity, and accordingly, it is also desirable that 2,6-dimethylnaphthalene (hereinafter, dimethylnaphthalene will be referred to as DMN regardless of the positions of substituent methyl groups) for forming the monomer component have a high purity. DMN has 10 isomers, and a highly pure 2,6-dimethylnaphthalene (preferably having a purity of 99% or more), which is mixed with substantially none of the

other 9 isomers, is preferably used.

As a method for manufacturing the 2,6-DMN described above, there may be mentioned a method of separating 2,6-DMN from an isomer mixture obtained by isomerizing 1,5-DMN which is formed by a reaction between orthoxylene and butadiene; a method comprising methylating naphthalene or methylnaphthalenes, subsequently isomerizing, and separating 2,6-DMN; and a method of separating 2,6-DMN from a tar or an oil fraction. However, the fractions and the products described above are each a mixture containing many types of DMN isomers in addition to 2,6-DMN, and hence, the 2,6-DMN must be separated from the mixture described above. However, since the boiling points of these DMN isomers are very close to each other, it has been difficult to separate a highly pure 2,6-DMN therefrom by distillation which is commonly used for separation/purification of organic compounds.

Accordingly, as a method for separating this 2,6-DMN, a crystallization method or an adsorption method has been proposed, and in addition to these methods mentioned above, for example, a method comprising forming a complex by using a certain organic compound, separating the complex, and decomposing this separated complex, and combinations of the methods described above have also been proposed. A cooling crystallization method is a method exploiting the property of 2,6-DMN having a highest melting point among the 10 types

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of DMN isomers, and since the cooling crystallization method is simple compared to the methods described above, this method can be used suitably as an industrial separation method. However, since it has been difficult to obtain a 2,6-DMN having a purity of 99% or more only by the cooling crystallization method, a process such as treatment using a solvent is generally used together therewith. For example, in Japanese Unexamined Patent Application Publication Nos. 48-5767 and 48-22449, and Japanese Examined Patent Application Publication No. 50-22553, a method has been disclosed in which after a mixture containing DMN isomers is crystallized by cooling, solid-liquid separation is performed by suction filtration, and the obtained solid component is dissolved in a solvent and is then crystallized by cooling. However, according to the related arts described above, a DMN mixture which primarily includes specific DMN isomers among the 10 types of isomers, such as 2,6-DMN, 1,6-DMN, and 1,5-DMN, which are easily isomerized to each other and are easily separated, is used as a feedstock, and a specific DMN mixture in which the content of 2,7-DMN which is difficult to separate from 2,6-DMN is limited to less than 5 mole percent (approximately equivalent to 5 wt) is used. A mixture containing DMN isomers obtained in a typical manufacturing process generally includes 5 wt% or more of 2,7-DMN, and hence, when

a mixture containing DMN isomers which includes 5 wt% or more of 2,7-DMN is used as a feedstock in accordance with the related art disclosed in the publications described above, it has been difficult to obtain a highly pure 2,6-DMN.

In view of the situations described above, the present invention was made, and an object of the present invention is to provide a method for manufacturing a highly pure 2,6-DMN even when a mixture containing DMN isomers which includes 5 wt% or more of 2,7-DMN is used as a feedstock.

Disclosure of Invention

A method for manufacturing 2,6-dimethylnaphthalene according to the present invention, which can solve the problems described above, comprises a step of performing cooling crystallization of a mixture containing dimethylnaphthalenes which includes 2,6-dimethylnaphthalene; a step of performing solid-liquid separation to obtain a solid component; and a washing step of washing the solid component using a solvent; wherein the solid-liquid separation performed after the cooling crystallization includes press filtration.

In addition, in a method for manufacturing a highly pure 2,6-dimethylnaphthalene according to the present invention comprising a step of performing cooling crystallization of a mixture containing dimethylnaphthalenes

which includes 2,6-dimethylnaphthalene, a step of performing solid-liquid separation to obtain a solid component, and a washing step of washing the solid component using a solvent, the washing step may be performed at least twice, and a part or the entirety of a mother liquor obtained in a second washing step or in a subsequent washing step may be used as a solvent in a washing step performed prior to the washing step at which the mother liquor is obtained.

In the method described above, the mixture containing dimethylnaphthalenes may be a mixture composed of dimethylnaphthalene isomers.

In the method of the present invention, the pressure of the press filtration is preferably 10 kg/cm² or more, and the press filtration is preferably performed using a tube press.

In addition, a mixture containing dimethylnaphthalenes, which includes 5 wt% or more of 2,7-dimethylnaphthalene, may be used as a feedstock, and the cooling crystallization may be performed for a mixture containing dimethylnaphthalenes which includes less than 25 wt% of 2,6-dimethylnaphthalene.

In both cases described above, a highly pure 2,6-dimethylnaphthalene can be manufactured.

In particular, when the washing step is performed for a solid component containing 80% or more of 2,6-dimethylnaphthalene by using a solvent, and subsequently,

solid-liquid separation and distillation are performed, a 2,6-dimethylnaphthalene having a high purity of 99% or more may be obtained.

According to the present invention, as the solvent used in the washing step, an aliphatic hydrocarbon and/or alicyclic hydrocarbon having 5 to 10 carbon atoms are preferably used.

Brief Description of the Drawings

Fig. 1 is a schematic diagram showing a typical example of a method of the present invention.

Fig. 2 includes views for illustrating a tube press method which is a typical example of press filtration.

Fig. 3 is a schematic diagram showing a typical example of a method of the present invention.

Best Mode for Carrying Out the Invention

In the case in which 2,6-DMN is separated by cooling crystallization and filtration from a mixture containing DMN isomers, which includes 5 wt% or more of 2,7-DMN, used as a feedstock, when the mixture containing DMN isomers is crystallized by cooling, the viscosity of a slurry of the mixture containing DMN isomers is increased. When the suction filtration is performed for this slurry in order to obtain 2,6-DMN, the separation efficiency from the mother

liquor is low, and hence, it is difficult to obtain a highly pure 2,6-DMN.

The inventors of the present invention carried out intensive research on the most significant subject that is to obtain a highly pure 2,6-DMN from a mixture containing DMN isomers, which includes 5 wt% or more of 2,7-DMN, used as a feedstock. As a result, it was discovered that when press filtration was incorporated in a solid-liquid separation process which was performed after cooling crystallization, a highly pure 2,6-DMN can be manufactured even when a mixture containing DMN isomers which includes 5 wt% or more of 2,7-DNM is used, whereby the present invention was made.

In the solid-liquid separation process, press filtration may be performed after a part of a mother liquor is removed by centrifugation from a slurry obtained by cooling crystallization, or may be preferably performed for a slurry obtained right after cooling crystallization.

As a press filtration method, for example, a tube press, a filter press, a plate press, a cage press, a belt press, a screw press, or a disc press method may be mentioned. Among these methods mentioned above, a method which can perform press filtration at a higher pressure is preferable when used for industrial mass production, and above all, the tube press method in which a high pressure of 100 kg/cm² or more

can be applied is preferably used.

The basic principle of the tube press method is as shown in Figs. 2(a) and (b). For example, a rubber-made cylinder 12 is concentrically provided inside a perforated cylinder 11 formed of a screen wound with a filter cloth, a slurry 13 is supplied into the annular gap formed between the two cylinders, and a high-pressure liquid is fed in the rubber-made cylinder so as to squeeze filtrate out of the perforated cylinder 11, whereby press filtration of the slurry 13 can be performed. The tube press method described above has been used in applications such as sludge treatment, but has not been used in the organic chemical industrial field since the rubber film made of raw rubber is dissolved in an organic solvent. However, as the rubber film described above, a fluorinated elastomer (polyfluorocarbon, for example, "Viton" manufactured by E. I. du Pont), which is insoluble in an organic solvent, has been developed in recent years, and hence, the tube press method can be used for the manufacturing method of the present invention.

As the mixture containing DMN isomers which is processed by cooling crystallization in the present invention, a mixture containing 5 wt% or more of 2,7-DMN may be used, and in addition, a mixture containing 10 wt% or more of 2,6-DMN is preferably used. When the content of 2,6-DMN in a feedstock less than 10 wt%, a mixture

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containing DMN isomers which includes 2,6-DMN at a low purity is preferably distilled so as to increase the concentration of 2,6-DMN to 10 wt% or more.

This mixture containing DMN isomers is processed by cooling crystallization to form a slurry, and the solid-liquid separation is performed for the slurry by press filtration, thereby yielding a highly pure 2,6-DMN. A crystal of the mixture containing DMN isomers, which is a solid component, is fed into a solvent for washing. A slurry containing the crystal obtained after washing is processed by solid-liquid separation using a common method such as centrifugation, and the solid thus separated is distilled for removing the solvent, thereby yielding a highly pure 2,6-DMN.

In the mixture containing DMN isomers obtained after cooling crystallization, in addition to 2,6-DMN, the other nine DMN isomers and other alkylnaphthalenes are contained, and most of the components are present in a liquid state (partly in a solid state). When these isomers are contained at high contents in the crystal obtained after solid-liquid separation, it is difficult to obtain a highly pure 2,6-DMN even after subsequent washing treatment. In order to obtain a highly pure 2,6-DMN, it is important to sufficiently perform solid-liquid separation by press filtration performed after cooling crystallization. Accordingly, the

higher the pressure during press filtration, the better the separation effect is. The pressure is preferably 10 kg/cm² or more, is more preferably 50 kg/cm² or more, and is even more preferably 80 kg/cm² or more.

By these cooling crystallization and press operation described above, the purity of 2,6-DMN in the mixture containing DMN isomers is preferably increased to 80% or more. Impurities in a liquid state present in the mixture containing DMN isomers, which contains 80% or more of 2,6-DMN, can be easily removed by washing using a solvent, and a high purity of 99% or more can be finally obtained. In the case in which the purity of 2,6-DMN cannot be 80% or more when the operation of the cooling crystallization followed by the press filtration is performed once (hereinafter, operation performed once may be referred to as "single stage" in some cases), the purity of 2,6-DMN is preferably increased to 80% or more by repeating the operation of the cooling crystallization followed by the press filtration (hereinafter, operation performed at least twice may be referred to as "multistage" in some cases). Since 2,6-DMN and 2,7-DMN have similar structures and properties to each other, their behaviors are also similar to each other, and hence, it is believed that most of impurities in a solid state present in the crystal of the mixture containing DMN isomers obtained after solid-liquid separation is 2,7-DMN.

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However, when the purity of 2,6-DMN is increased to 80% or more, and the content of 2,7-DMN in the crystal is decreased, alkynaphthalenes and the DMN isomers other than 2,6-DMN and 2,7-DMN, which are present in a liquid state, serve as a solvent to dissolve 2,7-DMN, and hence, most of this 2,7-DMN can be easily removed together with other impurities in a liquid state. On the other hand, when the purity of 2,6-DMN is less than 80%, the amount of a 2,7-DMN present in the crystal form is increased, and hence, it becomes difficult to remove 2,7-DMN only by washing. In addition, concerning a small amount of a 2,7-DMN in a solid state present in the crystal of the mixture containing DMN isomers obtained after solid-liquid separation, since the solubility of 2,7-DMN in a solvent is higher than that of 2,6-DMN, and 2,7-DMN is preferentially dissolved in a solvent, 2,7-DMN can be extracted from the crystal of the mixture containing DMN isomers by washing using a solvent.

The solvent used in washing treatment of the present invention is not specifically limited as long as it is in a liquid state at an operation temperature condition and is easily separated from DMN, and an aliphatic hydrocarbon or an alicyclic hydrocarbon is preferably used. The number of carbon atoms of an aliphatic hydrocarbon or an alicyclic hydrocarbon is preferably in the range of 5 to 10, and for example, hexane or octane may be mentioned.

The amount of a solvent on a weight basis is preferably at least one-third, is more preferably at least one times, is preferably at most 50 times, and is more preferably at most 5 times of the crystal of the mixture containing DMN isomers which primarily includes 2,6-DMN. In addition, the temperature range in washing treatment is preferably - 10°C or more, is more preferably 5°C or more, is preferably 45°C or less, and is more preferably 30°C or less.

As described above, by the washing treatment using a solvent, 2,7-DMN can be removed from the crystal of the mixture containing DMN isomers by extraction; however, when the content of 2,7-DMN is high, a 2,6-DMN having a high purity of 99% or more may not be obtained in some cases by performing the washing treatment once. In the case described above, a highly pure 2,6-DMN can be obtained by performing the washing treatment at least twice; however, when the washing treatment using a solvent is simply performed at least twice, the yield of 2,6-DMN may be significantly decreased in some cases. Accordingly, in order to prevent the decrease in yield described above, when the washing treatment is performed at least twice, as shown in Fig. 3 by way of example, a mother liquor obtained at a second or a subsequent washing treatment is used as a washing solvent at washing treatment prior to the washing treatment at which the mother liquor is obtained, whereby

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the purity of 2,6-DMN can be increased without decreasing the yield thereof.

Since a mother liquor obtained by washing treatment is a saturated solution of 2,6-DMN, 2,6-DMN is not further dissolved in the mother liquor when washing is performed using the solution described above. On the other hand, since impurities other than 2,6-DMN are not saturated, they are dissolved in this solution. Accordingly, when a mother liquor obtained by washing treatment is used as a washing solvent at washing treatment prior to the washing treatment at which the mother liquor is obtained, the purity can be increased without decreasing the yield of 2,6-DMN.

A solid component which is processed by the washing treatment described above at least twice is preferably obtained by cooling crystallization followed by the press filtration as described above; however, a solid component processed by solid-liquid separation using a method other than the press filtration may also be used.

In the present invention, since a crystal of the mixture containing DMN isomers obtained by press has a large block-shaped lump, when this crystal is dipped in a solvent for washing, it is difficult to remove impurities present at an inner side of the lump, and hence, the efficiency is low. Accordingly, the washing efficiency is preferably improved by providing a circulating pump outside a washing bath for

circulating a slurry therein and by pulverizing a cake to be washed using a wet-type pulverizer.

After a solvent separated from a highly pure 2,6-DMN by distillation or a solvent obtained by solid-liquid separation is processed by distillation so as to remove a solute, they may be reused for washing treatment.

Hereinafter, the present invention will be described with reference to figures; however, the present invention is not limited to the figures described below, and it is understood that every modification made in accordance with the description in this specification is within the scope of the present invention.

Fig. 1 is a schematic view of a typical example of the present invention in which a two-stage process of cooling crystallization and press filtration is shown.

After a mixture containing DMN isomers used as a feedstock is fed into a first cooling crystallization device 1 and is then cooled to a temperature below the melting point of 2,6-DMN, the mixture is supplied to a first press filtration device 2. A slurry containing a crystal obtained by the cooling is separated into a solution (mother liquor) and a solid component (crystal) by solid-liquid separation in this device, and the mother liquor is supplied outside the production line or is supplied to a former step by a pump P1. At the same time, the crystal is supplied to a

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second cooling crystallization device 3 and is then crystallized by cooling as in the first stage, and subsequently, solid-liquid separation was performed in a second press filtration device 4. The mother liquor obtained in this step is returned to the first cooling crystallization device 1 via a line L₁. In addition, a crystal obtained in this step is supplied to a washing bath 5 via a line L₂. The crystal is mixed with a solvent supplied into the washing bath 5 via a line L₃ and is washed by stirring. During washing by stirring, a slurry in the washing bath is supplied to a wet-type pulverizer 6 provided outside the washing bath, and a cake is pulverized and is then returned to the washing bath 5. After washing by stirring, the slurry is supplied to a subsequent solid-liquid separation step via a line L₄ (hereinafter, an example in which a centrifuge is used in a solid-liquid separation step is shown; however, another solid-liquid separation method may also be used). A 2,6-DMN cake, which is obtained by solid-liquid separation using a centrifuge 7, is supplied to a melting bath 8 via a line L₅, and after melting, the molten cake is supplied to a distillation tower 9 via a line L₆. The molten cake is separated into a solvent component and a product, i.e., a highly pure 2,6-DMN, in the distillation tower 9. The highly pure 2,6-DMN thus obtained is recovered as a product via a line L₇. At the

same time, the solvent is returned to the washing bath 5 via a line L₈ and the line L₃.

In addition, the solvent separated in the centrifuge 7 is supplied to a distillation tower 10 via a line L₉, and is then separated into a solvent component and a component of DMN mixture. The solvent component mentioned above is returned to the washing bath 5 via the line L₃, and the component of DMN mixture is returned to the first cooling crystallization device 1 via a line L₁₀.

Fig. 3 is a schematic view showing an example of a method for manufacturing 2,6-DMN in which washing treatment is performed at least twice, and as described above, the number of washing treatment may be increased or decreased in accordance with an object. A crystal (a solid component) obtained by solid-liquid separation using press filtration in a manner as described above is supplied to a washing bath 5A via a line L₂. The crystal described above is mixed with a solvent supplied into the washing bath 5A via a line L₃ and is washed by stirring. During washing by stirring, a slurry in the washing bath is supplied to a wet-type pulverizer 6 provided outside and is then returned to the washing bath 5A after a cake is pulverized. After washing by stirring, the slurry is supplied to a subsequent solid-liquid separation step (centrifuge 7) via a line L₄. A 2,6-DMN cake obtained by the solid-liquid separation using a

centrifuge 7A is supplied to a washing bath 5B, which is a second washing bath, via a line L₆.

In addition, a mother liquor obtained by separation using the centrifuge 7A is supplied to a distillation tower 9 via a line L₅ and is then separated into a solvent component and a low purity 2,6-DMN. The solvent component separated by the distillation tower 9 is supplied to the washing bath 5B, which is the second washing bath, from a line L₈ via a line L₁₁. Washing by stirring is performed in the washing bath 5B, as in the washing bath 5A, for the cake mixed with the solvent which is supplied via the line L₁₁. After washing by stirring, a slurry is supplied to a solid-liquid separation step. In this solid-liquid separation step, an example using a centrifuge is described; however, another solid-liquid separation method may also be used. A 2,6-DMN cake obtained by solid-liquid separation using a centrifuge 7B is supplied to a melting bath 8 via a line L₉, and after melting, the molten cake is supplied to a distillation tower 9B via a line L₁₀, so that the molten cake is separated into a solvent component and a highly pure 2,6-DMN. The solvent component described above is returned to the washing bath 5B via the line L₁₁.

The solvent obtained by separation using the centrifuge 7B is returned to the washing bath 5A via a line L₁₂. In addition, a highly pure 2,6-DMN is recovered as a product

from the distillation tower 9B via a line L₁₃.

Hereinafter, the present invention will be described in detail with reference to examples. However, the present invention is not limited to the examples described below, and it is understood that every modification made in accordance with the description in this specification is within the scope of the present invention. In the examples and comparative examples described below, "%" means "wt%".

EXAMPLE 1

A feedstock (mixture containing DMN isomers) having the composition shown in Table 1 was crystallized at 9°C and was then processed by press filtration at approximately 100 kg/cm², thereby obtaining a cake of the mixture containing DMN isomers having the composition shown in Table 1. Next, 100 g of this crystal of the mixture containing DMN isomers and 200 g of normal hexane were placed in a separable flask provided with a stirrer and were then stirred at 30°C for 1 hour. Subsequently, after a crystal was separated by suction filtration, 100 g of a pure solvent was poured over the crystal. When the crystal was analyzed using gas chromatography, it was found that a crystal having the composition shown in Table 1 was obtained. As "other impurities", there were methylnaphthalene, ethylnaphthalene, hydrocarbons having boiling points equivalent to those of

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the other DMN's, and the like.

Table 1

Composition (wt%)	Feedstock	Cake after Cooling crystallization/ Press Filtration	Filtrate after Cooling crystallization/ Press Filtration	Crystal after Washing (Example 1)
2,6-DMN	11.68	75.72	9.30	97.66
2,7-DMN	12.03	10.54	12.09	2.34
Other DMN isomers	26.23	6.01	6.01	Below detection limit
Other Impurities	50.06	7.93	7.93	Below detection limit
Total	100.00	100.00	100.00	100.00

The yield with respect to 2,6-DMN in the cake before washing was 65.71%. It was understood that 2,6-DMN could be obtained with a high yield according to the present invention.

EXAMPLE 2

A cake of a mixture containing DMN isomers having the composition shown in Table 2 and a crystal formed therefrom after washing were obtained in a manner equivalent to that in example 1 except that a feedstock (mixture containing DMN isomers) having the composition shown in Table 2 was crystallized at 15°C in a first stage and was then crystallized at 70°C in a second stage, and that press filtration was performed at approximately 100 kg/cm² after each cooling crystallization mentioned above.

Table 2

The yield with respect to 2,6-DMN in the cake before washing was 65.05%. It was understood that 2,6-DMN could be obtained with a high yield according to the present invention.

EXAMPLE 3

A cake of a mixture containing DMN isomers having the composition shown in Table 3 and a crystal formed therefrom after washing were obtained in a manner equivalent to that in example 1 except that a feedstock (mixture containing DMN isomers) having the composition shown in Table 3 was crystallized at 3°C and was then processed by solid-liquid separation using a centrifuge in a first stage, and in a second stage, a crystal was precipitated at 65.8°C and was then processed by press filtration using a tube press at an operation pressure of 100 kg/cm². As the tube press in this example, TPS-1 provided with temperature control function manufactured by Ashizawa Co., Ltd. was used in which the filtration area was 0.45 m² and the inside volume was 17 liter.

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Table 3

Composition (wt%)	Feedstock	Cake after First Cooling Crystallization	Filtrate after First Cooling Crystallization	Cake after Second Cooling Crystallization	Filtrate after Second Cooling Crystallization	Crystal after Washing (Example 3)
2,6-DMN	10.98	41.1	8.0	95.56	35.73	100.00
2,7-DMN	12.56	14.7	12.1	2.73	16.73	Below Detection Limit
Other Impurities	76.50	44.3	79.9	1.71	47.50	Below Detection Limit
Total	100.00	100.00	100.00	100.00	100.00	100.00

The yield with respect to 2,6-DMN in the cake before washing was 77.24%. It was understood that 2,6-DMN could be obtained with a high yield according to the present invention.

EXAMPLE 4

A cake of a mixture containing DMN isomers having the composition shown in Table 4 and a crystal formed therefrom after washing were obtained in a manner equivalent to that in example 1 except that a feedstock (mixture containing DMN isomers) having the composition shown in Table 4 was crystallized at 29°C and was then processed by press filtration at approximately 100 kg/cm².

Table 4

Composition (wt%)	Feedstock	Cake after Cooling crystallization/ Press Filtration	Filtrate after Cooling crystallization/ Press Filtration	Crystal after Washing (Example 4)
2,6-DMN	20.46	83.93	14.19	99.10
2,7-DMN	10.53	4.74	11.72	0.78
Other DMN isomers	22.45	4.32	25.49	0.12
Other Impurities	46.56	7.01	48.60	Below detection limit
Total	100.00	100.00	100.00	100.00

The yield with respect to 2,6-DMN in the cake before washing was 66.66%. It was understood that 2,6-DMN could be

obtained with a high yield according to the present invention.

Example 5

Multistage washing including return of a mother liquor was performed as described below.

- (1) A feedstock (mixture containing DMN isomers) having the composition shown in Table 5 was crystallized at 32°C and was then processed by press filtration at approximately 100 kg/cm², thereby obtaining a cake of the mixture containing DMN isomers having the composition shown in Table 5.

(2) Next, 545 g of this crystal of the mixture containing DMN isomers and 660 g of normal hexane were placed in a separable flask provided with a stirrer and were then stirred at 30°C for 1 hour. Subsequently, the crystal was separated by press filtration, and the mother liquor was recovered.

(3) Next, by using 165 g of the crystal of the mixture containing DMN isomers obtained in the above (1) and 660 g of the mother liquor obtained in the above (2), washing treatment was performed in a manner equivalent to that performed in the above (2) except that the temperature was set to 35°C. Subsequently, the crystal was separated by centrifugation.

(4) In addition, by using 128 g of the crystal obtained in

the above (3) and 411 g of normal hexane, washing treatment was performed in a manner equivalent to that performed in the above (2). Subsequently, the crystal was separated by press filtration, and when the crystal thus obtained was analyzed using gas chromatography, it was found that the crystal had the composition shown in Table 5. The yield with respect to 2,6-DMN in the cake before washing was 45%.

Table 5

Composition (wt%)	Feedstock	Cake after Cooling Crystallization	Filtrate after Cooling Crystallization	Crystal after First Washing ((2) in This Example)	Crystal after Second Washing ((4) in This Example)
2,6-DMN	21.84	81.77	15.39	97.76	99.05
2,7-DMN	18.76	7.76	18.49	1.98	0.87
Other DMN isomers	57.46	9.93	61.18	0.26	0.08
Other Impurities	4.43	0.54	4.94	Below Detection Limit	Below Detection Limit
Total	100.00	100.00	100.00	100.00	100.00

Comparative Example 1

A cake of the mixture containing DMN isomers having the composition shown in Table 6 and a crystal formed therefrom after washing were obtained in a manner equivalent to that in example 1 except that the a feedstock (mixture containing DMN isomers) having the composition shown in Table 6 was crystallized at 26°C and was then processed by suction

filtration while approximately 2 kg/cm² was applied.

Table 6

Composition (wt%)	Feedstock	Cake after Cooling crystallization/ Suction Filtration	Filtrate after Cooling crystallization/ Suction Filtration
2,6-DMN	21.95	37.74	15.09
2,7-DMN	10.45	7.50	11.24
Other Impurities	67.50	54.76	73.67
Total	100.00	100.00	100.00

The purity of 2,6-DMN in the crystal obtained after washing was less than 70%.

Industrial Applicability

As has thus been described, according to the method of the present invention, even when a DMN mixture containing 5 wt% or more of 2,7-DMN is used as a feedstock, a highly pure 2,6-DMN can be manufactured, and in addition, even when a DMN mixture containing less than 25 wt% of 2,6-DMN is processed by cooling crystallization, a highly pure 2,6-DMN can be manufactured. In particular, when a solid component containing 80% or more of 2,6-DMN is washed using a solvent and was then processed by solid-liquid separation and distillation, a 2,6-DMN having a high purity of 99% or more can be obtained.

CLAIMS

1. A method for manufacturing highly pure 2,6-dimethylnaphthalene comprising: a step of performing cooling crystallization of a mixture containing dimethylnaphthalenes which includes 2,6-dimethylnaphthalene; a step of performing solid-liquid separation to obtain a solid component; and a washing step of washing the solid component using a solvent; wherein the solid-liquid separation performed after the cooling crystallization includes press filtration.

2. A method for manufacturing highly pure 2,6-dimethylnaphthalene comprising: a step of performing cooling crystallization of a mixture containing dimethylnaphthalenes which includes 2,6-dimethylnaphthalene; a step of performing solid-liquid separation to obtain a solid component; and a washing step of washing the solid component using a solvent; wherein the washing step is performed at least twice, and a part or the entirety of a mother liquor obtained in a second washing step or in a subsequent washing step is used as a solvent in a washing step performed prior to the washing step at which the mother liquor is obtained.

3. The manufacturing method according to one of Claims 1 and 2, wherein the mixture containing dimethylnaphthalenes

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is a mixture composed of dimethylnaphthalene isomers.

4. The manufacturing method according to one of Claims 1 to 3, wherein the press filtration is performed at a pressure of 10 kg/cm² or more.

5. The manufacturing method according to one of Claims 1 to 4, wherein the mixture containing dimethylnaphthalenes used as a feedstock includes 5 wt% or more of 2,7-dimethylnaphthalene.

6. The manufacturing method according to one of Claims 1 to 5, wherein the cooling crystallization is performed for a mixture containing dimethylnaphthalenes which includes less than 25 wt% of 2,6-dimethylnaphthalene.

7. The manufacturing method according to one of Claims 1 to 6, wherein the washing step is performed for a solid component containing 80% or more of 2,6-dimethylnaphthalene using a solvent, and further comprising steps of performing solid-liquid separation and distillation after the washing step, whereby a 2,6-dimethylnaphthalene having a high purity of 99% or more is obtained.

8. The manufacturing method according to one of Claims 1

to 7, wherein the solvent used in the washing step is an aliphatic hydrocarbon and/or alicyclic hydrocarbon having 5 to 10 carbon atoms.

9. The manufacturing method according to one of Claims 1 to 8, wherein the press filtration is performed using a tube press.

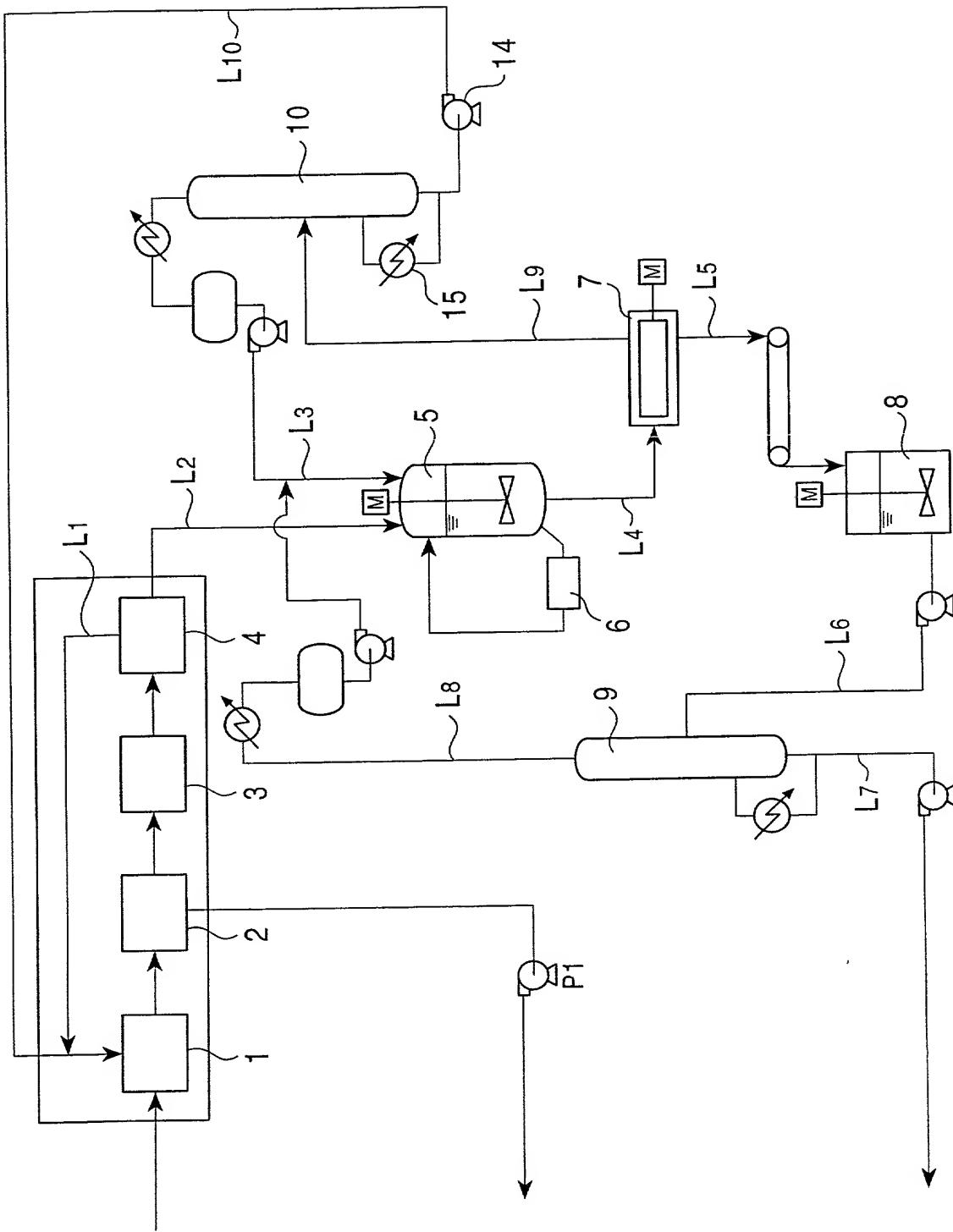
ABSTRACT

An object of the present invention is to provide a method for manufacturing 2,6-DMN, in which even when a mixture containing DMN isomers which includes 5 wt% or more of 2,7-DMN is used, a highly pure 2,6-DMN can be obtained. The method for manufacturing the highly pure 2,6-dimethylnaphthalene of the present invention comprises performing cooling crystallization of a mixture containing dimethylnaphthalenes which includes 2,6-dimethylnaphthalene, performing solid-liquid separation to obtain a solid component, and washing the solid component using a solvent, wherein the solid-liquid separation performed after the cooling crystallization includes press filtration. In the present invention, the pressure of the press filtration is preferably 10 kg/cm² or more, and according to the method of the present invention, even when a DMN mixture containing 5 wt% or more of 2,7-DMN is used as a feedstock, a highly pure 2,6-DMN can be manufactured, and in addition, even when a DMN mixture containing less than 25 wt% of 2,6-DMN is processed by cooling crystallization, a highly pure 2,6-DMN can be manufactured.

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FIG. 1



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FIG. 2A

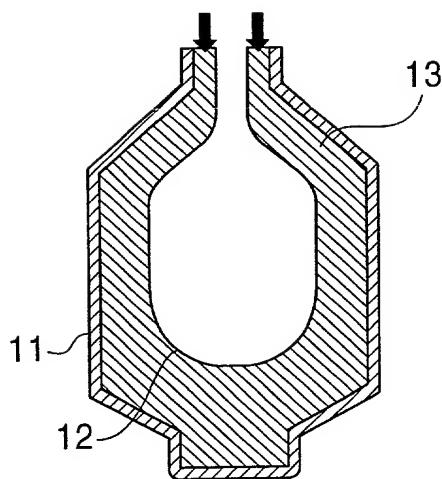
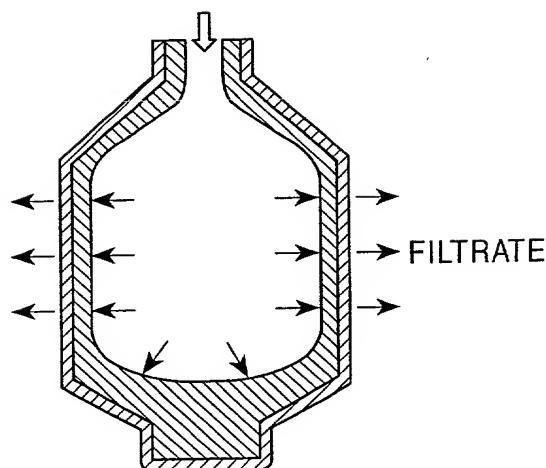
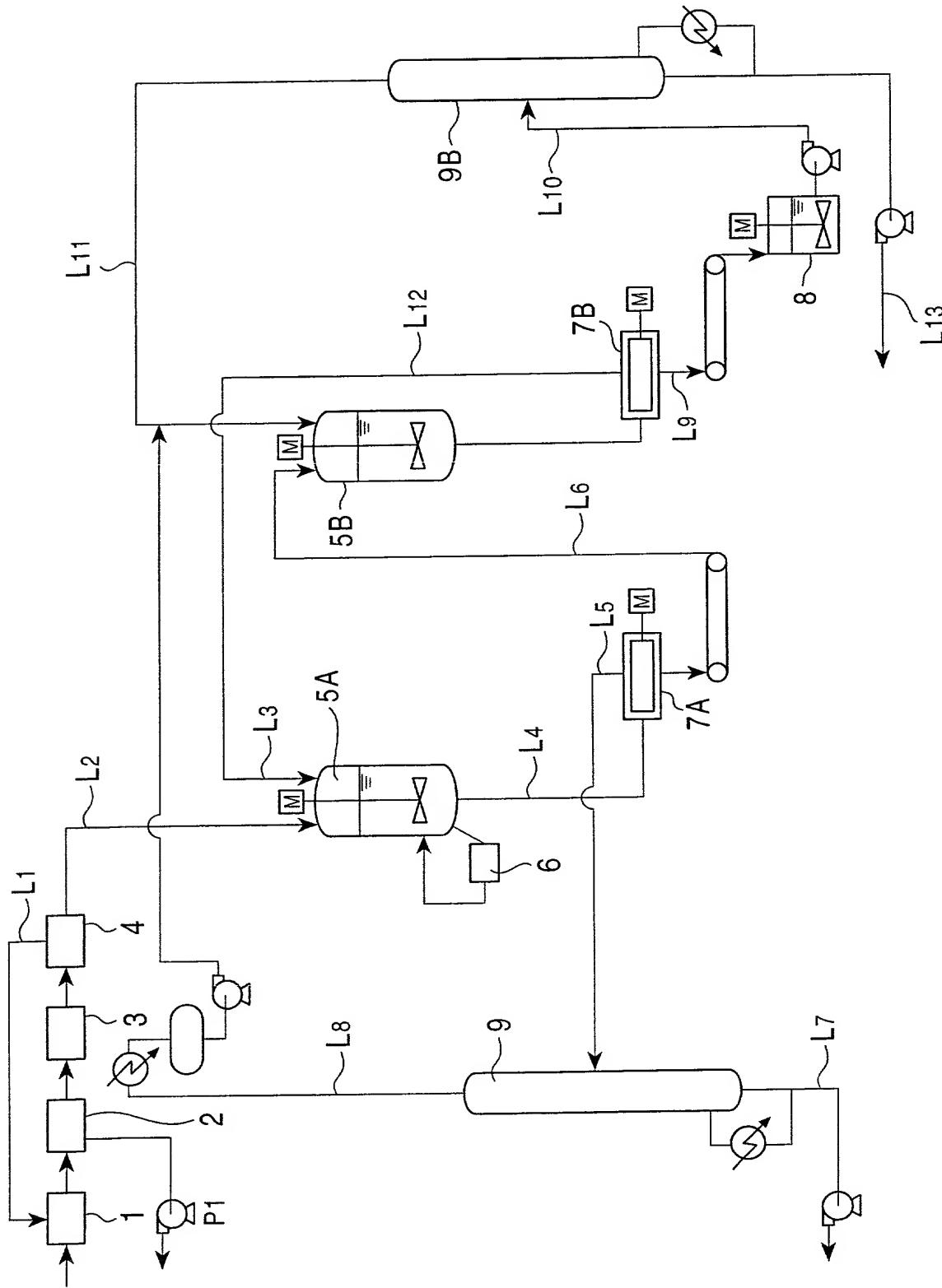


FIG. 2B



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FIG. 3



Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者（下記の名称が複数の場合）であると信じています。

上記発明の明細書は、

- 本書に添付されています。
 ____月____日に提出され、米国出願番号または特許協定条約国際出願番号を_____とし、
（該当する場合）_____に訂正されました。

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled.

METHOD FOR MANUFACTURING 2,6-
DIMETHYLNAPHTHALENE

the specification of which

- is attached hereto.
 was filed on _____
as United States Application Number or
PCT International Application Number
_____ and was amended on
_____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulation, Section 1.56.

Japanese Language Declaration

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私は、米国法典第 35 編 119 条 (a) - (d) 項又は 365 条 (b) 項に基づき下記の、米国以外の国の少なくとも一ヵ国を指定している特許協力条約 365 (a) 項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

Prior Foreign Application(s)

外国での先行出願

Number 番号	Country 国名
0011-246299	JP

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Month/Day/Year Filed 出願年月日	Priority Claimed 優先権主張
August 31 1999	YES

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(出願番号)

(Filing Date)
(出願日)

PCT/JP00/05844

August 30,2000

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(出願番号)

(Filing Date)
(出願日)

I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

(Status: Patented, Pending, Abandoned)
(現況 : 特許許可済、係属中、放棄済)

Patented Pending Abandoned

I hereby declare that all statements made herein of my own knowledge are true and that all statement made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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委任状：私は下記の発明者として、本出願に関する一切の手続きを米特許商標局に対して遂行する弁理士または代理人として、下記の者を指名いたします。

(弁護士、または代理人の指名及び登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: (list name and registration number)

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(第三以降の共同発明者についても同様に記載し、署名すること)。

Supply similar information and signature for third and subsequent joint inventors.)

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第三共同発明者 3-00	Full name of third joint inventor, if any Takeharu TANAKA
第三共同発明者の署名 日付 ✓ 田中文雄	Third Inventor's signature Date Feb. 6, 2002
住所	Residence c/o Kobe Corporate Research Laboratories in Kobe Steel, Ltd 5-5, Takatsukada 1-chome, Nishi ku, Kobe-shi, Hyogo 651-2271 Japan
国籍	Citizenship JP Jpx
郵便の宛先	Post Office Address SAME AS ABOVE

第四共同発明者 4-00	Full name of fourth joint inventor, if any Masahiro MOTOYUKI
第四共同発明者の署名 日付 ✓ Masahiro Motoyuki	Fourth Inventor's signature Date Feb. 6, 2002
住所	Residence c/o Osaka Branch in Kobe Steel, Ltd 1-3, Bingo machi 4-chome, Chuo-ku Osaka shi, Osaka 541-0051 Japan
国籍	Citizenship JP Jpx
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第五共同発明者 5-00	Full name of fifth joint inventor, if any Shingo YOSHIDA
第五共同発明者の署名 日付 ✓ 石田 神吾	Fifth Inventor's signature Date Feb. 6, 2002
住所	Residence c/o Osaka Branch in Kobe Steel, Ltd 1-3, Bingo machi 4-chome, Chuo-ku, Osaka-shi, Osaka 541-0051 Japan
国籍	Citizenship JP Jpx
郵便の宛先	Post Office Address SAME AS ABOVE

第六共同発明者	Full name of sixth joint inventor, if any
第六共同発明者の署名 日付	Sixth Inventor's signature Date
住所	Residence
国籍	Citizenship
郵便の宛先	Post Office Address

(第六またはそれ以降の共同発明者についても同様な
情報および署名を提供すること。)

(Supply similar information and signature for sixth and
subsequent joint inventors.)